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VOPROSY RADIOAKTIVNOSTI

(Problems of Radioactivity)

by

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ABSTRACT

The reports presented by American, Japanese and Soviet scientists to the Tenth Pacific Congress at Honolulu, Hawaii are discussed. The quantity of Sr^{90} and Cs^{137}, and other radium isotopes, in various regions and at various depths of the Pacific Ocean is outlined, disclosing the degree of ocean water pollution by radioactive waste materials resulting from atom bomb explosions. The various methods used in the determination of water pollution are compared and analyzed, exemplifying the processes by tables, graphs and formulae.

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PROBLEMS OF RADIOACTIVITY

As is known, the problems of investigations of radioactivity in the World Ocean have become very actual during the last decade. After a series of tests of various types of nuclear weapons and also due to the use of seas and oceans by several foreign countries as a gigantic reservoir for the disposition of radioactive waste of the atom industry, the radioactivity of the World Ocean can be presented as a sum of two components — the artificial and natural radioactivity.

Although the artificial radioactivity, caused by the fall-outs and the disposition of radioactive products into oceans, constitutes only part of the natural phenomenon, one must account for their effect, considering the fact that the basic elements are Cs^{137} and Sr^{90}.

Inasmuch as the greater portion of tests of the powerful nuclear and thermonuclear weapons was carried out in the central part of the Pacific Ocean, some of the countries such as Japan, U.S.A. and also several others have shown a special interest in the investigations of radioactivity in the Pacific waters. As early as the Ninth Pacific Congress in Bangkok a large number of participants had devoted their reports to problems of measuring radioactivity. The importance of the problem determined the conduction of a special symposium named the "Radioactive indicators in oceanography". Such a theme made it possible to unify the most diverse themes within the framework of one symposium. Here, the problems of methods and apparatus for measuring the radioactivity were discussed; also important problems of oceanology that have to be solved at investigations were elucidated.

As is known, the investigation of radioactivity of seas comprises two aspects. On the one hand, the investigations enable us to determine the degree of water pollution in a given area. On the other hand, the investigations offer an opportunity to observe large-scale processes by artificial and natural utilization of radioactive isotopes as indicators. Such radioactive marks can be specifically introduced into the composition of ocean waters. In addition, the radioactive matter that has been formed as a result of nuclear and thermonuclear explosions, as well as the matters of cosmogenic origin, fall out over the ocean surface. By investigating their distribution with depth, one can draw conclusions relative to the intensity of exchange processes in the ocean.

The trend of the Congress, naturally, determined also the participants for the symposium. The greater part of the scientists were representatives of the U. S. A. Not a small number represented Japan. Several participants were from other countries. The main problems mentioned in reports and discussions in the Congress were as follows: 1) the methods of measurements and the development of new instruments; 2) the organization of investigations of radioactivity; 3) data on the degree of pollution in specified areas of the Pacific Ocean; 4) the determination of the intensity of diffusion by means of radioactive indicators; 5) the problems of the age of water.

The Methods of Measurements and Instruments /458

At the present time, the most widely used method of determining the radioactivity of sea water is a method based on chemical liberation of a given element from water samples having a quantity of 50 to 200 ℓ. The beta-calculation is made by means of the Geiger-Müller computers. The gamma-calculation is carried out by a multichannel gamma-spectrometer with the use of a spectrometric sodium iodide scintillator (as a rule). It is with the aid of such methods that the results cited in a report by Miyake [10] were obtained. The author bases his views on a method of determining Sr^{90} and Cs^{137} which has already been discussed by him [11, 12]. In the given case, Sr^{90} was liberated from water in the form of strontium nitrate, whereas Cs^{137} was deposited with the ammonium phosphomolybdate. The measurement of Sr^{90} beta-activity was made by the Geiger-Müller computer at 0.5 imp/min. The results were compared with data obtained by the measurement of activity of Y^{90} types. The measurement of Cs^{137} gamma-activity was carried out by a gamma-spectrometer with a scintillator of sodium iodide (5 x 5 cm) within the energy range from 0.52 to 0.80 MeV. For comparison, the Cs^{137} solutions of standard activity were investigated.

Folsom [2], in his investigations of the activity of Cs^{137} on the basis of the same principles, used, however, a different method of concentration. Refusing to utilize the cobalt sodium nitrate, which was used by him earlier [3], he achieved the concentration by adding nickel sulfate and potassium ferrocyanide. According to the author, this method is more acceptable for the investigation of tropical waters, having, in addition, other advantages [4].

In line with the description of standard methods an attempt was made to construct an apparatus for direct measurements of artificial radioactivity in the water. The report by Proctor et al [13] is devoted to

the description of an instrument designed for determining the gamma-radioactivity. The instrument represents a gamma-spectrometer, which makes it possible to determine the elements on the basis of characteristic lines of their gamma-spectra. The scintillator, photomultiplier, as well as the transformatory part of the instrument, were placed into a metal container which was submerged to a depth of 80 m in the Puget Sound (Seattle, U. S. A. area). In the field investigations, conducted from aboard a small trawler, the signal was t ismitted by a cable to the vessel where, with the aid of a single-channel analyzer, the integral spectrum of radiation was investigated. The application of a single-channel analyzer did not enable the authors to carry out any measurements except for the determination of intensity of cosmic radiation at various depths (the results agree sufficiently well with data by other authors [5, 16]). As could be expected, it was established that the background of the instrument conditioned by cosmic rays decreases considerably in the upper 10-meter layer. Thus, the measurement of activity at small depths (1-3 m) is considerably aggravated during the motion of the vessel. Due to the limited extent of background and its limited variation with depth, all the investigations beneath 10 m can be carried out in any weather conditions.

A considerably more complete investigation of the instrument was carried out by the authors in a laboratory. Creating in a tank with a volume of several m^3 solutions of a definite concentration for many radioactive isotopes, they determined their gamma-spectra with the aid of a 256-channel analyzer with the energy range to 3 MeV. Various types of scintillators were investigated (sodium iodide, plastic, liquid). The influence of the size of scintillator on the effectiveness of recording has been elucidated. As could be expected, the most effective was the scintillator of the sodium iodide having the maximum size (13 x 10 cm). Average qualities passed the scintillator having a size of 7.5 x 7.5 cm. Thus, with a further improvement of instruments, attempts will be made to determine the activity of waste and sea waters.

Organization of Marine Investigations of Radioactivity /459

In connection with the fact that a considerable increase in the degree of ocean pollution has been lately observed (according to Folsom, the concentration of radiostrontium in the coastal waters of the U. S. A., in the La Jolla area, has increased twice during the brief period from the spring of 1960 to the spring of 1961), the U. S. A. and Japan have organized numerous expeditions, which, being very mobile, make it possible to obtain exhaustive material on the most diverse areas of the

ocean. From June 1960 to July 1961 the Scripps Institution of Oceanography alone conducted four expeditions to various parts of the Pacific Ocean. By the time the Congress was opened, two expeditions were in the ocean and one was to return soon. The objective of such types of investigations is to obtain the values of radioactivity on the ocean surface at a maximum possible number of points. The radioactivity of more than 120 samples of surface water was determined during the period. The investigation of bottom waters was not, evidently, carried out by the expeditions organized by the U. S. A.

Analogous investigations were conducted by Japan, mainly in the northwestern portion of the Pacific Ocean. During the same time, the investigations conducted by Japanese scientists in the vessel "Riofu-Maru" (1200 t), which belongs to the Japanese Meteorological Agency, made it possible to obtain data on radioactivity at depths to 8000 m and to conclude on the intensity of penetration of radioactive pollution at ocean depths.

The Degree of Ocean Pollution

As is known, the ocean waters have a natural radioactivity. This radioactivity is caused by the fact that natural radioactive isotopes of several elements, K^{40} primarily, are found in the composition of sea water. An idea on the elements causing the natural radioactivity of sea water is obtained by a summarized table included in Proctor's report to the Congress [13]. This table is based on data by Revell et al [16], as well as by Lowder and Solon [9]. The half-lives of the elements have been taken from Seaborg's tables [15].

The Content of Radioisotopes in Sea Water [13]

Isotope	Concentration, g/ml	Activity μc/ml	Half-life, years	Type of decomposition and energy, MeV
K^{40}	$4,5\times10^{-3}$	$3,3\times10^{-7}$	$1,25\times10^{9}$	89% β, 11% εγ 1,46
H^{3}	8×10^{-14}	$6,8\times10^{-9}$	12,3	β
Rb^{87}	$8,4\times10^{-3}$	$5,9\times10^{-8}$	50	100% β
U^{238}	$1,5\times10^{-3}$	$2,0\times10^{-8}$	$4,51\times10^{9}$	α, β, weak γ*
Ra^{226}	$0,2\times3\times10^{-10}$	$0,54—8,1\times10^{-10}$	1662	α, β, Pronounced γ* 0,05—2,4
C^{14}	4×10^{-11}	$1,9\times10^{-10}$	5568	β
U^{235}	$1,5\times10^{-5}$	$8,1\times10^{-11}$	$7,1\times10^{8}$	α, β, weak γ*
Th^{232}	$1,0\times10^{-5}$	$5,4\times10^{-11}$	$1,39\times10^{10}$	α, very weak γ*
La^{138}	$2,5\times10^{-7}$	2×10^{-15}	$1,0\times10^{11}$	β 0,54; 0,81; 1,44

*Including the activity of daughter products.

Also, the data collected by two reporters concerning the magnitudes of artificial radioactivity in various areas of the Pacific Ocean were discussed at the symposium. The reporters were Folsom [2] and Miyake with associates [10]. Especial interest and lively debates were created by Miyake's report. He presented the results of determination of Sr^{90} and Cs^{137} in the waters of the northwestern portion of the Pacific Ocean. According to these data, obtained in 1960, the concentration of the elements exceeded 30 to 40 times the concentration calculated by Folsom for other areas of the ocean. Miyake found a considerable concentration of the elements in deep waters to 5000 m (they made up 0.1 $\mu\mu c/\ell$). Beneath the depth, the branching radioactivity was not observed. It is interesting to note that the average ratio Cs^{137}/Sr^{90} in sea water was 1.2, which is considerably below the quantity that is usually observed in fall-outs (from 1.7 to 2.8). The

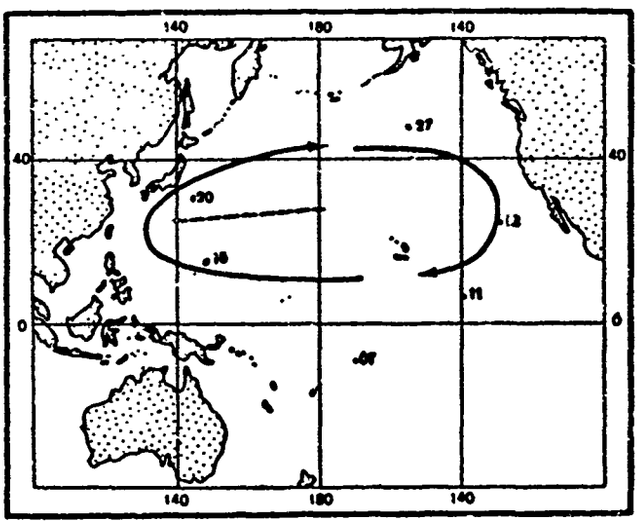

Fig. 1. Distribution of Cs^{137} in the surface waters of the Pacific Ocean, $\mu\mu c/\ell$ [2].

difference is obviously explained by the fact that there are substantial differences in the periods of semidecomposition of their predecessors Xe^{137} and Kr^{90} (3.8 min and 33 sec, respectively). The greater portion of Xe^{137} can be left in the stratosphere prior to the formation of Cs^{137}. At the same time, the major portion of Sr^{90} may fall out on the sea surface due to a shorter half-life of Kr^{90}. The figures calculated by Miyake for the Sr^{90} and Cs^{137} concentrations in surface waters (3.1 $\mu\mu c/\ell$ and 4.8 $\mu\mu c/\ell$, respectively) differ

considerably from the results presented by Folsom, who leads at present the investigation of oceanic radioactivity in the U. S. A. (fig. 1). According to him, the northern waters of the Pacific Ocean, moving in an eastward direction, contain somewhat more radiocesium than the waters of other areas. Between November 1959 and September 1961, the radioactivity of the waters was periodically determined in the La Jolla area. An increase in the concentration from 0.1 in the spring of 1960 to 0.2 in 1961 was observed. Nevertheless, Folsom expresses doubt that at the present time the average value has substantially exceeded the latter magnitude for any area of the Pacific Ocean.

Because of such differences in data, the conduction of a joint American-Japanese expedition is planned, which will present a possibility of comparing the methods and elucidate the causes for the differences.

Determination of Intensity of Diffusion With the Aid of Radioactive Indicators

The investigations conducted by Miyake and associates in 1958-1960 enabled them not only to find figures characterizing the degree of pollution in the northwestern part of the Pacific Ocean at various depths to 8000 m, but also to determine on the basis of the figures the coefficients of vertical turbulent diffusion. They calculated the coefficient with the aid of the known formula used for the solution of the diffusion equation for a momentary current source:

$$C = \frac{C_0}{\sqrt{\pi D t}} \exp\left[-\frac{z^2}{4Dt}\right],$$

in which z denotes the depth; t is time; C_0 the total quantity of radioisotopes introduced in the surface layer in time interval $t = 0$; C denotes the concentration of radioisotopes at $t = t$ and $z = z$. The authors determined the coefficient of turbulent diffusion in the ocean beneath the discontinuity layer. Assuming that $t = 3$ years for layer 200-4000 m, the determined coefficient D was 200. Accounting for it, they found the variation with time of the vertical distribution of nonactive material introduced into the upper 200-m layer. The time needed for the creation of a practically homogeneous distribution of admixture to a depth of 4000 m (when the concentration in the depth constitutes 99% of concentration at surface) was appraised to be 700 years. As is known, Harley [6] determined the coefficients of vertical

turbulent diffusion on the basis of the distribution of beta-activity in layer 100-1000 m. For the tropical areas of the Pacific Ocean the figure was D = 40. Miyake explains the difference of results by increase of coefficient D, with depth as a consequence of the decrease of stability.

It need be mentioned that although the results are very interesting, the use of a simplified calculation method (without accounting for vertical speeds, without giving the currents of radioactivity on the surface, the use of the equation of momentary current source) makes it possible to form only a general idea and qualitative evaluation of the process.

The calculations of Kolesnikov et al [7], which were presented to the Congress, are based on a more complete scheme. The distribution with depth of radioactive elements contained in fall-outs was based on calculations accounting for the radioactivity on the ocean surface, for the vertical components of mean current speeds, and the T 1/2 of each element. The coefficients of turbulent diffusion and the vertical velocities were determined by measurements conducted in the Atlantic Ocean during the expedition of the survey vessel "Mikhail Lomonosov". The calculation is based on a three-layer model: 1) in the intermixed layer lying over the discontinuity layer, the coefficient of turbulent diffusion is constant and the vertical velocity equals zero; 2) in the intermediate layer the coefficient increases according to a linear law, and the vertical velocity is constant; 3) in the bottom layer the coefficient decreases according to a linear law, and the vertical velocity equals zero.

The resultant calculation demonstrates that there is a considerable difference in the character of distribution of radioactive elements with depth for the high latitudinal areas where the water masses sink, and for tropical areas where they rise. This difference points out /462
the important significance of vertical velocities in the general transport of radioactivity.

Also the report of Ozmidov [13], devoted to the problems of horizontal turbulent diffusion, outlined the problem of distribution of radioactivity. By using the known law of "power $^{4/3}$" $K_\ell(\ell) = k\ell^{4/3}$, which characterizes the variation of the coefficient of turbulent diffusion with the scale of the phenomenon, the author arrives at the equation of horizontal turbulent diffusion in this form:

$$\frac{dq}{\partial t} = \frac{d}{dx}\left(k\sqrt{x^2+y^2}^{4/3}\frac{\partial q}{\partial x}\right) + \frac{d}{dy}\left(k\sqrt{x^2+y^2}^{4/3}\frac{dq}{dy}\right),$$

where q denotes the concentration of diffusing admixture; x, y denote the Descartes coordinates of observation point; k denotes the coefficient of proportionality. The solution of this equation in polar coordinates

$$q(r,t)=\frac{Q_1}{6\pi(4/3k)^{3}+3}\exp\left[-\frac{r^{4/3}}{4/3\,kt}\right]$$

(Q_1 being the linear strength of the source) enabled the author to calculate the propagation of radioactive substances and, without considering the coefficient of turbulent diffusion, to compare them with data on the propagation of radioactive substances formed as a result of nuclear explosions at the Bikini Atoll ("Operation Baker", July 25, 1946). In accordance with the existing evaluations, the coefficient of proportionality k equals 0.0136 $cm^{2/3}$ sec^{-1}. The resultant formula describes considerably better the distribution of radioactivity along the horizontal and is in considerably better agreement with experiments than the definition of the Gauss curve processes used before. This is especially true of the central parts of radioactive spots.

Problems of the Age of Water

The problems of the age of water is very important when determining the processes of intermixing occurring in the ocean. For example, their solution aids in determining the speed of propagation of radioactive pollution in a given area.

A report by Rafter [14] was devoted to the method of measurements and results of determining the age by radiocarbon C^{14}. He had employed a pronounced method; however, the data concerning the determination of the age of water along the coast of New Zealand were rather broad. A noteworthy fact is the abundance of observation stations along the entire coast of New Zealand. This made it possible to obtain vast material on the radioactivity of the sea and atmosphere in the given area. Despite the generally known deficiencies of the method (too great a T 1/2 of C^{14}, changes in its concentration in nature as a result of industrial effects, as well as in connection with the conduction of thermonuclear tests), the report defends the method. Besides, it is necessary to note that the author soberly appraises the deficiencies of this method. As a curiosity, Rafter cited the result of determining the age of a fish. It appeared to equal 500 (!) years.

Koczy presented in his report very exhaustive data on the determination of the age of waters in the World Ocean [8]. The data were obtained by measuring the heavy natural radioactive elements in sea water. The author discussed thoroughly the differences of the methods in the determination of age by means of heavy isotopes. The radium method was considered as the most rational method. Of great interest were the data on the distribution of radium in the World Ocean and the age of water determined by Koczy (fig. 2). The reporter has carried out a detailed analysis of errors in the measurements, citing the following figures that characterize the scattering of results concerning the

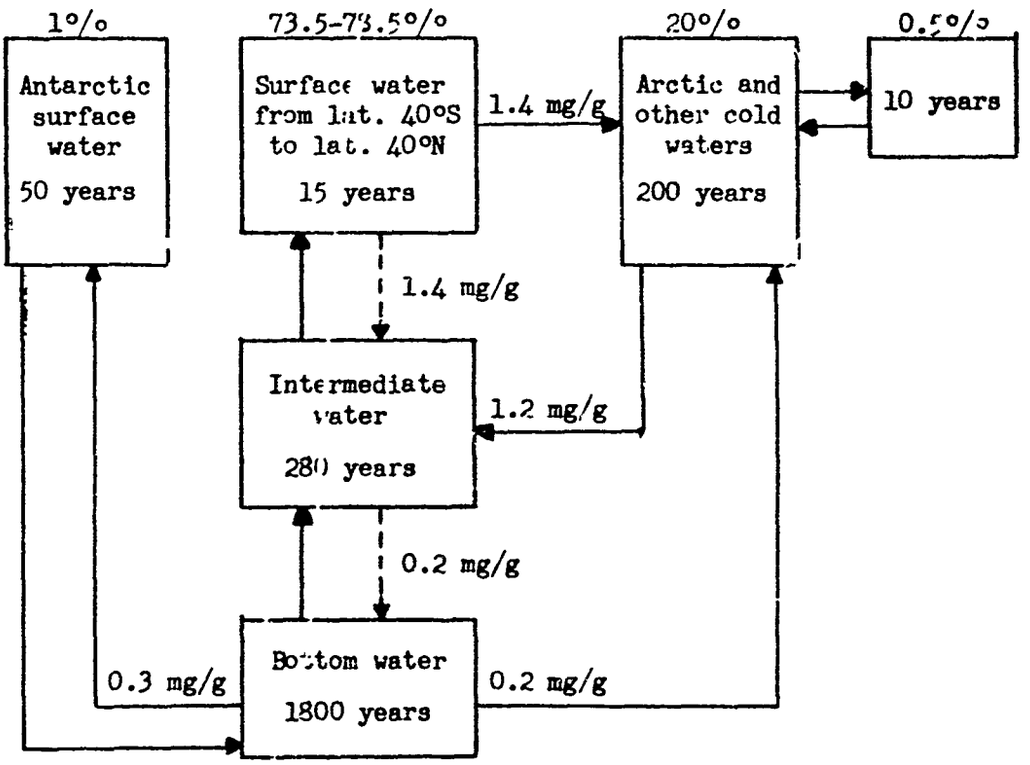

Fig. 2. Table of the age of ocean waters [8].

determination of radium concentration at various depths: at the bottom, (9—16) x 10^{-17} g/ml; at the surface, (4—9) x 10^{-17} g/ml.

Appraising the significance of the symposium as a whole, it need be noted that the extensive discussion of problems covering radioactivity, planned and prepared in advance, enabled the scientists of many

countries to discuss the problems in this field of oceanography. Undoubtedly, a further discussion of the problems will be of great value.

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